CHAPTER III

EXPERIMENTAL DETAILS

3.1 An Introduction to Molecular Beam Epitaxy

Molecular beam epitaxy (MBE) is a process for growing thin epitaxial films of a wide variety of materials. The growth process is well controlled by using the modern tools of surface analysis to obtain a real time surface structure and its environment. In this process, beams of atoms or molecules in an ultra-high vacuum environment are incident upon a heated crystal that has previously been processed to produce an atomically clean surface. The arriving constituent atoms form a crystalline layer in registry with the substrate, i.e., an epitaxial film. The composition of films can be rapidly changed, producing crystalline interfaces that are almost atomically abrupt. Because of the cleanliness of the growth environment and because of the precise control over composition, MBE structures closely approximate the idealized models used in solid state theory

Fig. 3.1 shows a schematic front view of a basic MBE growth chamber. A thin, crystalline substrate wafer is mounted on a heater block such that it can be brought to face the source ovens used to evaporate the constituent atoms or molecules. Mechanical shutters driven from outside the vacuum chamber are used to switch the beams on and off. Because of the extensive use of chilled walls surrounding the source ovens and the substrate, the beams make essentially a single pass through the chamber before condensing on the cold walls, and the background pressure in the system remains very low. This preserves the purity of the growing film and at the same time allows the reflection high energy electron diffraction (RHEED) gun to operate without damage from corrosive reaction with residual vapors. The RHEED system provides a diffraction pattern on a phosphor-coated window that is indicative of the ordering of the substrate surface. Thus the observer can immediately see the effect on film crystallinity due to changes in the growth conditions, e.g., exposing the surface to the source beams or changing beam intensity or substrate temperature.

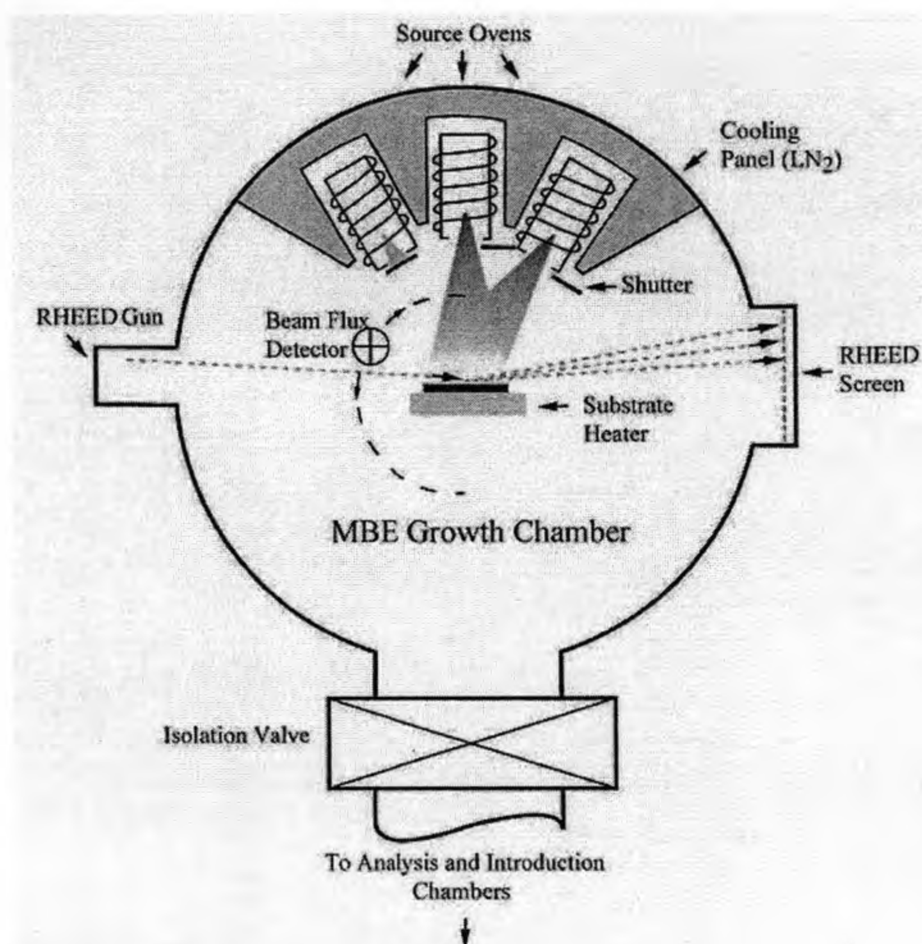

Figure 3.1: A schematic of MBE growth chamber showing the essential growth sources, shutters, beam flux detector and the RHEED system for monitoring structure during growth.

All samples in this work were grown in a conventional RIBER 32P solid source MBE. The MBE system consists of three chambers, i.e., introduction chamber, transfer chamber, and growth chamber. A schematic drawing of the modified III-V MBE growth chamber is shown in Figure 3.2 These 3 chambers are separated by isolation gate valves. In introduction chambers, heaters used for a heat treatment process of the substrate are established. Introduction chamber, transfer chamber, and growth chamber are generally kept in ultra high vacuum condition by titanium sublimation pumps, and an ion pumps. The pressure inside the chambers and beam flux pressure are measured by ion gauges. RHEED and quadrapole mass spectrometer is used to acquire the real time information of growth process and growth chamber conditions. The substrate holder can be rotated by a

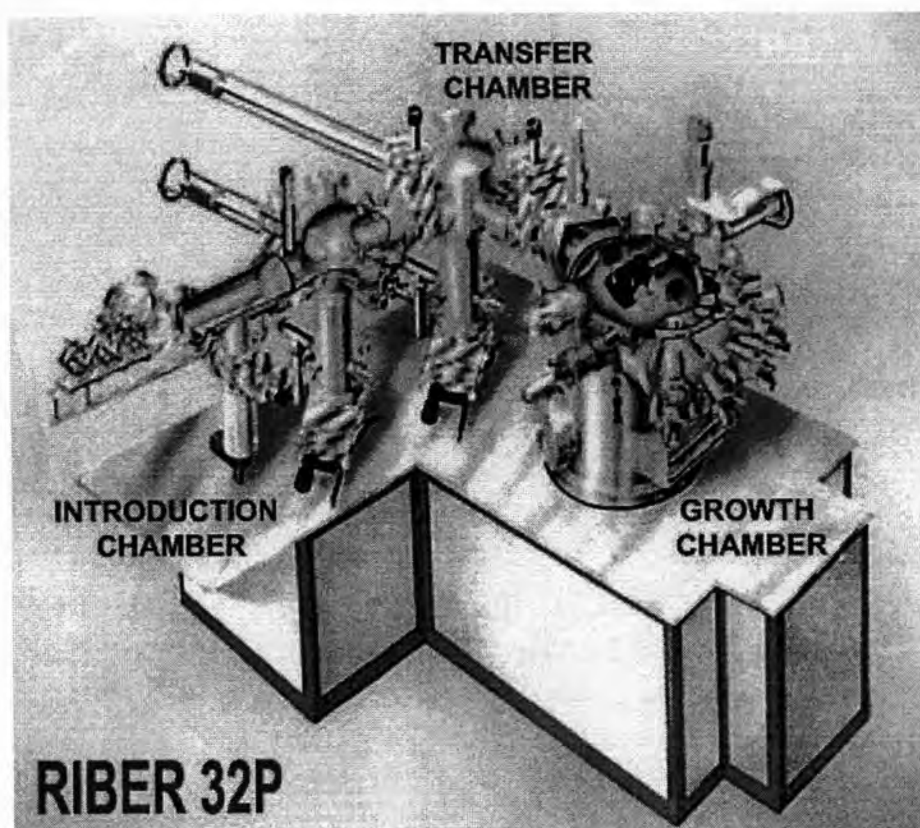

Figure 3.2: The modified RIBER 32P MBE consists of three chambers, i.e., introduction chamber, transfer chamber, and growth chamber [31].

speed-adjustable motor to achieve uniform growth.

3.2 Reflection High Energy Electron Diffraction

RHEED is a essential tool for characterizing the surface structure and morphology. An electron beam generated by electron gun is incident on a crystal surface at a grazing angle of a few degrees. In general operation energy of electron beam is used in range between 10 and 20 keV. At the crystal surface, there is a scattering process and diffracted beams leave the crystal and strike to a phosphor screen. RHEED is extremely sensitive to the atomic structure of the first few planes of a crystal lattice. The interpretation of electron interference pattern on phosphor screen is complicated. As a simple case, the two-dimensional array of lattice point produce a streak pattern on the intersection of Edward sphere and reciprocal lattice rods (in Fig. 3.4) whose intensity is in a kind of Laue function of reciprocal lattice.

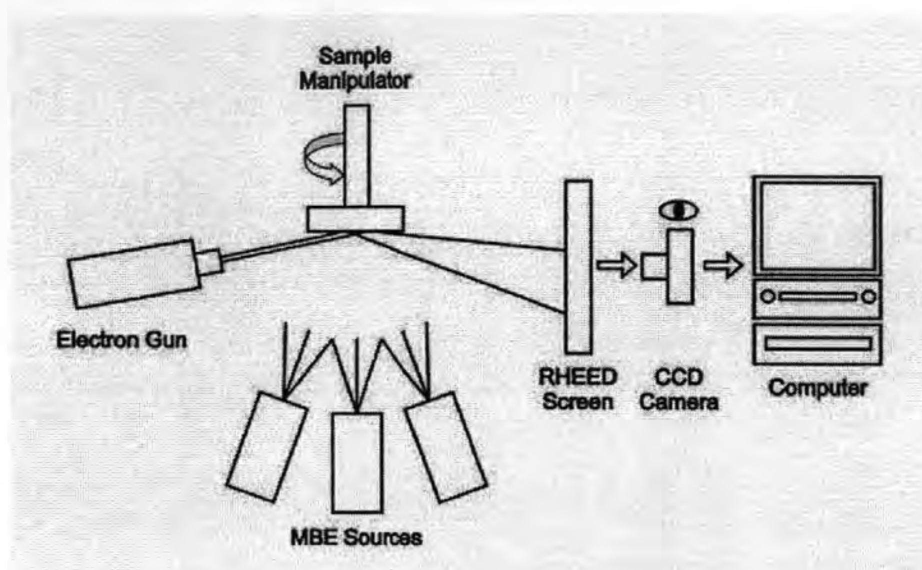

Figure 3.3: The schematic of RHEED system which consist of electron gun, RHEED screen and CCD camera connected to a computer.

3.3 Growth Rate Calibration

3.3.1 Beam Equivalent Pressure (BEP)

This measurement is dependent on the factors such as the geometry of the system and ionization efficiency of the material being measured. But for a given system and material, BEP is proportional to the flux at the sample surface and the growth rate. Unlike other techniques, a BEP measurement does not require a growth of epitaxial layers. Therefore, we use this method after already knowing the relation between growth rates and pressures of each material by the other methods whereby we deduce that the system's condition is not changed if the chamber is not leaked out.

3.3.2 RHEED Intensity Oscillation

A layer-by-layer growth, the growth of each atomic layer is largely completed before the next begins. The arriving atoms first nucleate in 2-D islands if steps are not present on the smooth surface. Subsequently, arriving atoms can migrate to the existing step edges to complete the monolayer and return the surface to a smooth condition. Thus the surface cycles between smooth and atomically rough, with a period corresponding to the time to complete a monolayer of growth. The rougher surface causes more diffuse scattering of

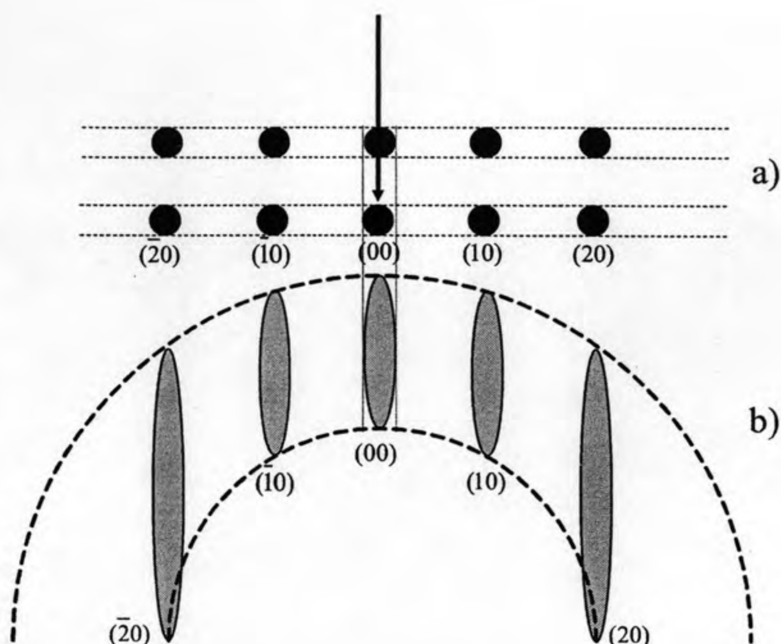

Figure 3.4: a) Reciprocal lattice of two-dimensional array of lattice points, b) RHEED construction for a) [32]

the RHEED beam, leading to a lower intensity of the diffracted beams. The oscillation frequency thus corresponds to the monolayer growth rate. In this work, this method is used for GaAs calibrations. The calibrations was done at substrate temperature of 580°C which is typically used for the growth of GaAs.

3.3.3 RHEED Pattern Transition of Strain Relaxing

In SK mode, after grown layers reach the critical thickness, 3D islands are formed. Thus, RHEED pattern changes from streak to chevron. By utilization of knowledge of

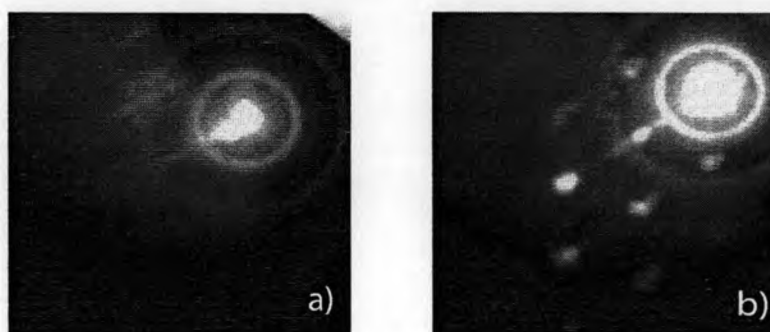

Figure 3.5: a) RHEED Pattern of GaAs at 580 °C, b) RHEED Pattern of 1.8ML InAs QDs

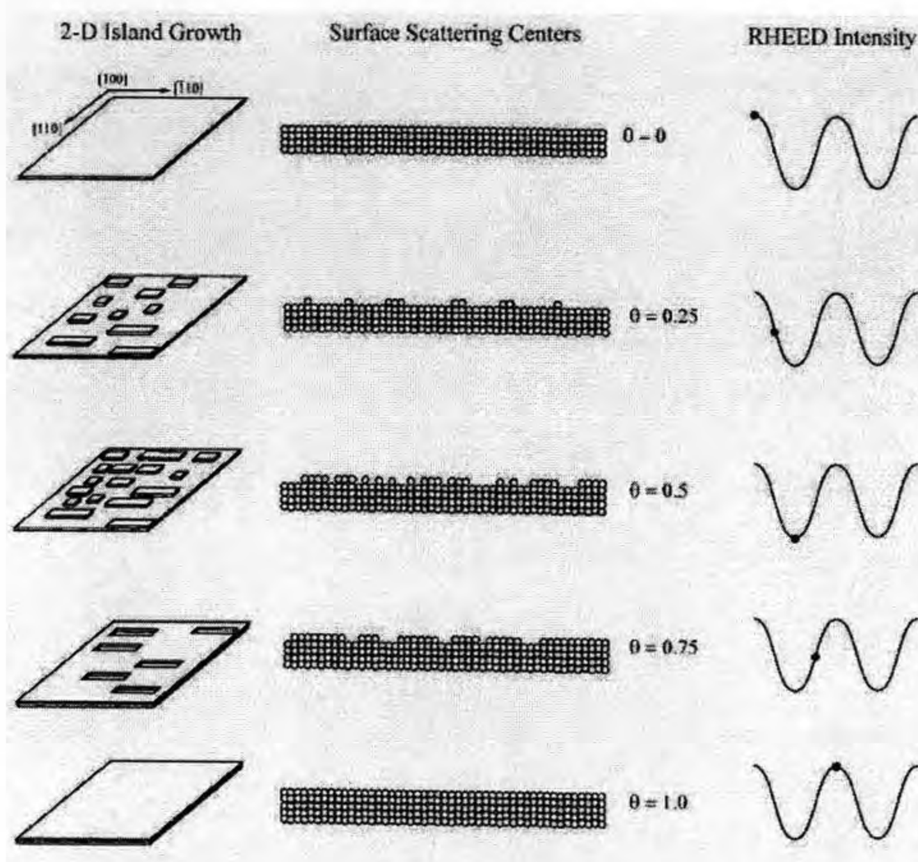

Figure 3.6: A Schematic diagram of the correlation of surface coverage of 2-D clusters with idealized RHEED oscillation.

critical thickness (1.7 ML for InAs on GaAs(001)), growth rate can be calculated from the deposition time which was spent when pattern transition occured. The calibrations must be done under unity sticking conditions. The substrate temperatures during calibration of InAs was hence held at 450°C because InAs desorption dramatically increases at higher temperature.

3.4 Substrate Temperature Calibration

For GaAs(001), a surface structure depends on temperature of substrate and As pressure at surface. At the typical growth temperature, GaAs surface structure arrange themselves into $c(2 \times 4)$ reconstruction. When the substrate temperature decrease, the RHEED pattern along the [011] direction changes from $c(2 \times 4)$ to $c(2 \times 2)$ pattern. In this work, transition temperature is 500 °C which is used as a reference point by calibration

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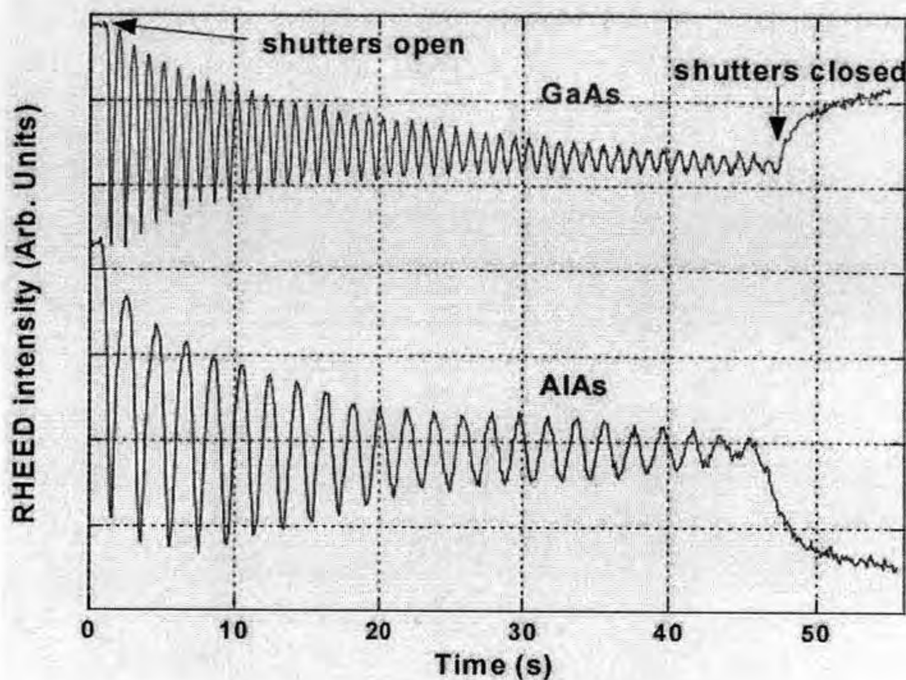

Figure 3.7: RHEED intensity oscillations of the specular spot during GaAs (top) and AlAs (bottom) growth.

under As overpressure of 8×10^{-6} Torr. In order to calibrate substrate temperature, we recorded four temperature values from thermocouple behind the block by the time substrate temperature was reduced from growth temperature of GaAs-buffer; (T_1) when secondary line at the centre of two main lines disappeared. (T_2) when secondary line at the centre of two main lines appeared but the others disappeared. In other word, $c(2 \times 2)$ pattern begun at T_2. By the time a substrate temperature was ramped up, we also recorded the temperature (T_3) when $c(2 \times 4)$ pattern terminated or secondary line at the centre of two main lines disappeared, and the substrate temperature (T_4) when the three secondary lines of $c(2 \times 4)$ pattern entirely appeared. The average value of the four recorded temperature $((T_1 + T_2 + T_3 + T_4)/4)$ was used as a reference substrate temperature of 500 °C.

3.5 Sample Characterization

3.5.1 Atomic Force Microscope (AFM)

In this work, the AFM images are performed by using SEIKO SPA 400-AFM in tapping mode. The AFM consists of a cantilever with a sharp tip at its end that is used to

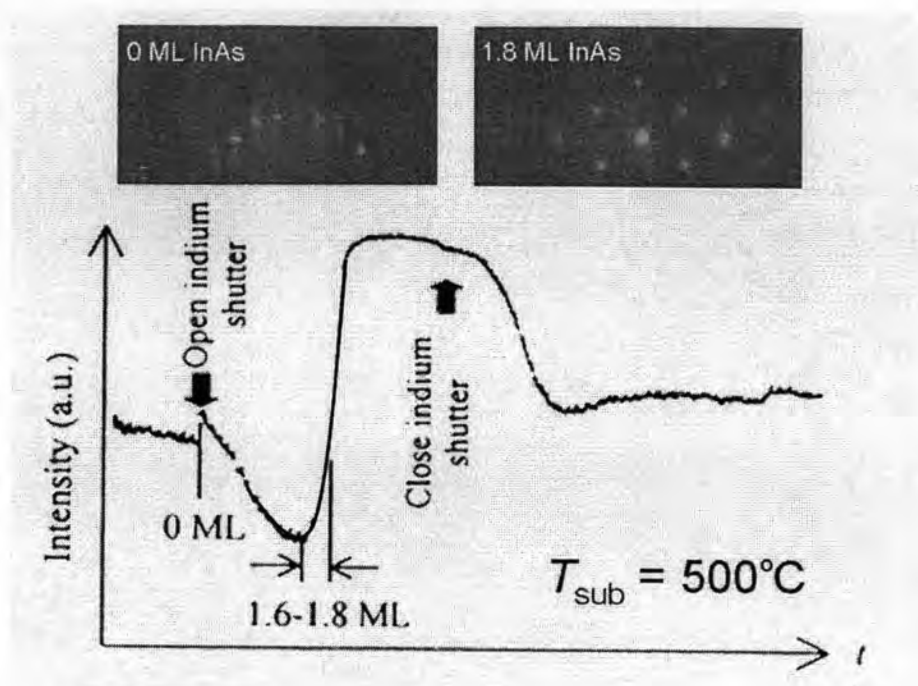

Figure 3.8: RHEED intensity as a function of InAs deposition time showing the increasing of intensity when the InAs layer has relaxed to form QDs

scan the specimen surface. The cantilever is typically silicon or silicon nitride with a tip radius of curvature on the order of nanometers. When the tip is brought into proximity of a sample surface, forces between the tip and the sample lead to a deflection of the cantilever. The deflection is measured using a laser spot reflected from the top of the cantilever into an array of photodiodes. The sample is mounted on a piezoelectric tube, that can move the sample in the z direction for maintaining a constant force, and the x and y directions for scanning the sample.

3.5.2 Photoluminescence (PL)

A schematic of the PL measurement system is shown in Figure 3.11. The samples were excited by the an Ar^+ laser at wavelength of 488 nm. The laser beam was chopped into pulse signal and focused by focal lens before it is incident on the samples. Light emitting from the samples was focused into a monochomator. The intensity of monochromatic light was measured by photomultiplier and signal was amplified and filtered by a lock-in amplifier. A PL spectra was programmatically scanned by adjusting wavelength selection of monochromator. For polarization-dependence measurement, the PL system

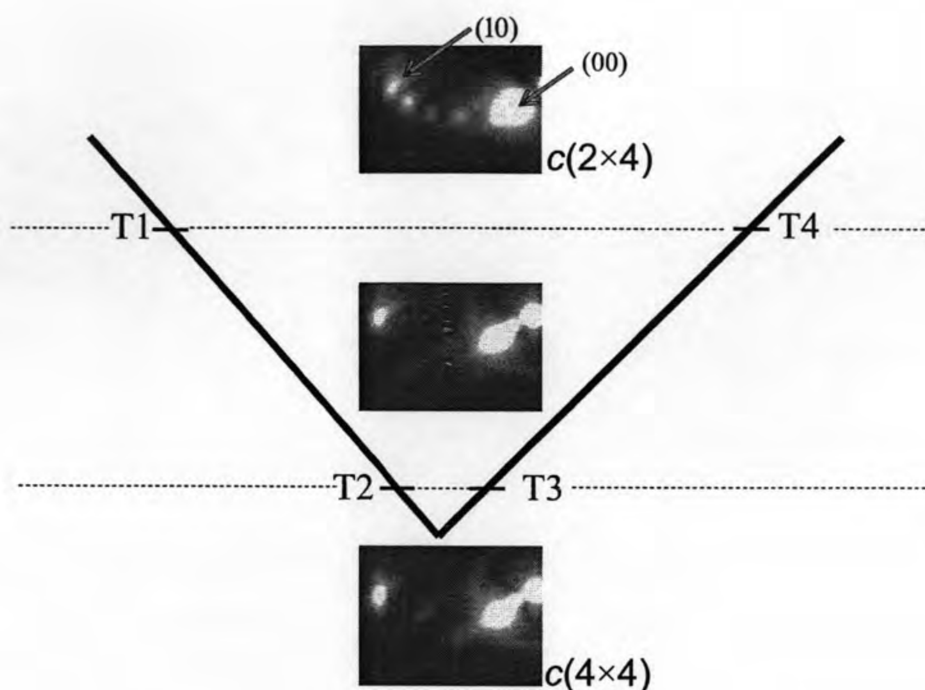

Figure 3.9: Temperature calibration process of GaAs substrate by RHEED pattern transition.

was modified. A half wave plate was placed in front of samples to adjust the polarized direction of incident laser and polarizer was placed between samples and a monochromator. The polarized direction of laser was adjusted to obtain the highest excitation. After that, the polarizer was retarded to determine the highest intensity and set that direction to be angle of zero as a reference angle($\theta = 0$). The polarizer was retarded to each angle for scanning PL spectra or recording intensity of PL peak. The light passing through polarizer at θ angle is corresponding to Malus's law. For linear polarized light, intensity is $I = I_0 \cos^2(\theta)$.

3.6 Samples Preparation

Substrates were mounted on a molybdenum block with In glue and loaded into load-lock chamber. In order to transfer the substrates to introduction chamber, load-lock chamber was pumped by a diaphragm pump and sorption pumps. They was heated at 450°C for an hour under vacuum pressure lower than 10^{-8} Torr before were ready to do a growth process. In order to prepare epi-ready surface, the substrate temperature

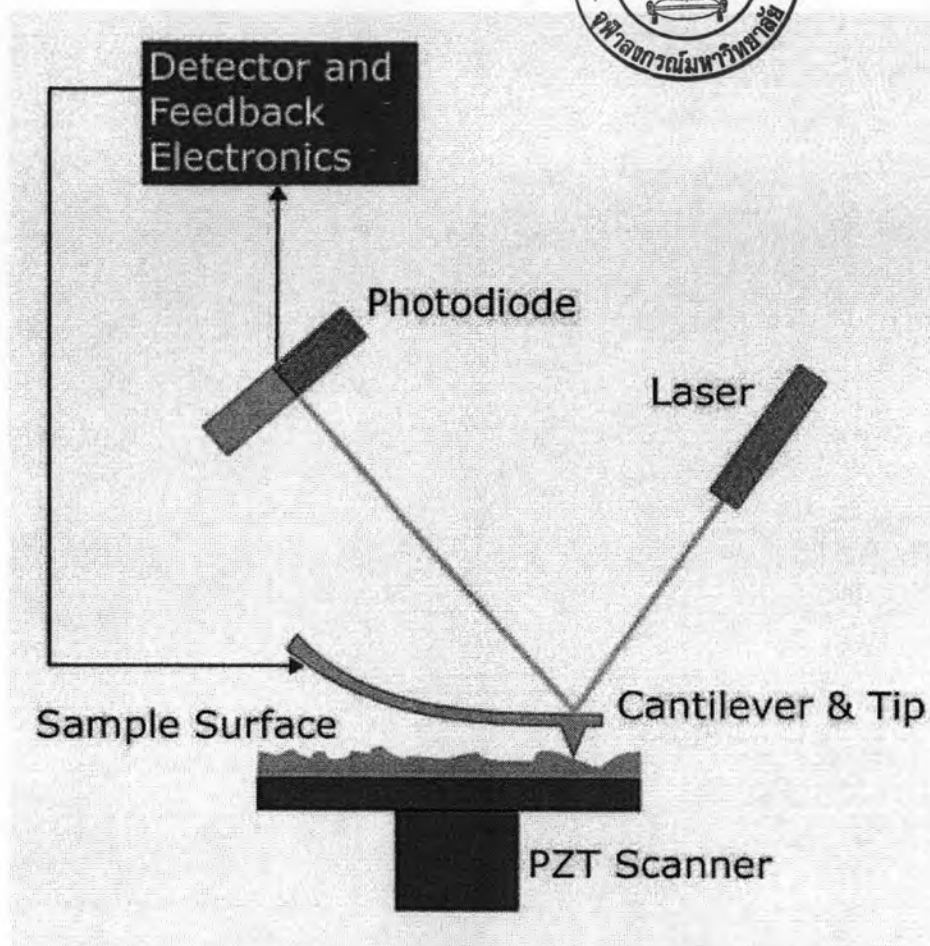

Figure 3.10: A schematic diagram of AFM

was ramped up in the growth chamber. When it reached to 300°C, the shutter of As cell was opened to leave them under As pressure about 8×10^{-6} Torr. the substrate temperature continued to be increased, until a diffraction pattern appears, showing that the oxide desorbed from the surface. This temperature monitored by thermocouple is used as a reference temperature of 580°C for growth of buffer layer. After RHEED shows the desorption of oxide, the sample temperature was increased more 20°C and was hold, until amount of oxide in the chamber monitored by quadrapole mass spectrometer decrease to acceptable value. A 300 nm of GaAs buffer was grown at 580°C to prepare a smooth surface for growth of nanostructures.

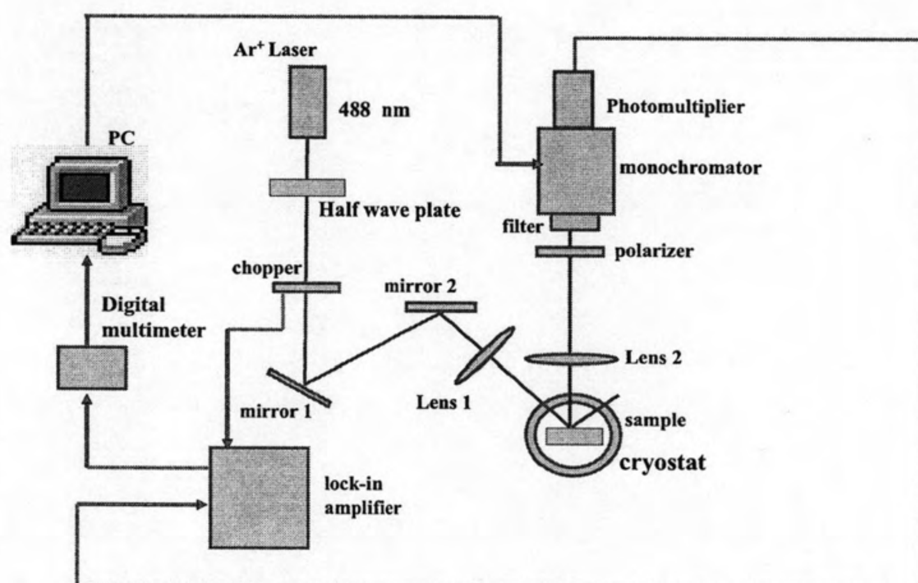

Figure 3.11: A schematic diagram of the polarization-dependence of PL measurement system